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(54) LOW CALORIE FAT SUBSTITUTES

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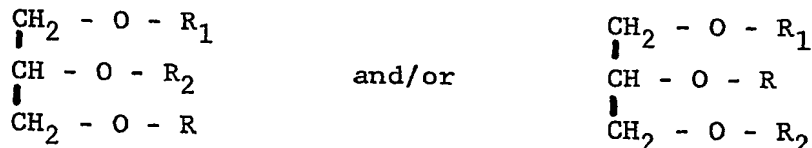
LOW CALORIE FAT SUBSTITUTES

Abstract of the Disclosure:

Glycerol diethers and glycerol monoester diethers are used as substitutes for conventional fats (glycerol esters) in food products.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. A method of providing a low calorie food composition, said method comprising formulating a food product to contain, at least as a portion of its lipid content, dialkyl ether compounds having the formula



wherein R_1 and R_2 are the same or different, saturated or unsaturated, straight or branched, alkyl hydrocarbon chains of from 8 to 30 carbon atoms; and R is either hydrogen or $-\text{C}-\text{R}_3$ where R_3 is an acyclic hydrocarbon chain, saturated or unsaturated straight or branched, of from 3 to 21 carbon atoms.

2. The method of claim 1 wherein R_1 and R_2 are alkyl chains of from 12 to 24 carbon atoms.

3. The method of claim 1 wherein R is hydrogen.

4. The method of claim 1 wherein the food product is fabricated from protein, carbohydrates and said dialkyl ether compounds.

5. The method of claim 1 wherein the composition is formulated by replacing at least a portion of the normal fat content of a food product with said dialkyl ethers.

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PATENT AGENTS



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Specification:

This invention relates to the use of fat-like materials in food products, and more particularly relates to the use of glycerol dialkyl ethers in food products.

Foods are usually divided into three classes: protein, carbohydrate, and lipid (fats). In general, of the three classes of food, fats or lipids contain the highest energy level per unit weight. The most common lipids are the glycerides, e.g., monoglycerides, diglycerides and triglycerides. The lipids most commonly found in foods are the triglycerides. Triglycerides are commonly known as fats or oils, depending upon their melting point. The term "fats" will be used hereafter to denote triglycerides regardless of their melting point. In addition to providing nutritional requirements, fats play an extremely important role in food technology, providing characteristic lubricity, texture, appearance, flavor, satiety, as well as other functional roles. Fat-containing foods, from which the normal fat component has been removed, are usually unacceptable due to poor texture, flavor, structure, and lubricity.

Fats provide food energy of about 9 kilocalories per gram, while proteins and carbohydrates provide about 4 kilocalories per gram. In the normal diet, fat contributes about 20% to 40% of the daily intake of calories. Thus, for those people who wish to control their caloric intake, a reduction of fat in their diet produces a greater overall decrease of calories than a reduction in proteins or carbohydrates of equal weight. However, reducing the consumption of fats the diet is difficult due to the non-availability of low fat food sources, and due to the decreased palatability of such foods, and also due to the attraction of most people for fat-containing foods. For the above reasons, there is a



real need for foods having a low fat content but high acceptability. If a low calorie substitute for fat could be found, which substitute met the functional requirements of fat, then low calorie diet foods having acceptable organoleptic properties could be prepared.

It is therefore a primary objective of the present invention to provide a group of compounds that can be used as fat substitutes.

It is a further objective of the present invention to provide low calorie food compositions containing such fat substitutes.

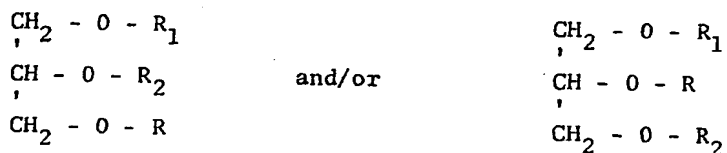
10 The present invention provides dialkyl glycerol ethers and glycerol monoester diethers having functional properties similar to those of conventional fats.

The dialkyl glycerol ethers are preferably not significantly absorbed in the alimentary canal.

The present invention is also directed at providing novel blends of dialkyl glycerol ethers, which blends have functional properties similar to those of conventional fats.

20 Furthermore, the present invention is concerned with the provision of diether compounds and blends that are not broken down by naturally occurring enzymes found in the digestive tract, and therefore are not absorbed in the digestive tract to any significant degree.

According to the present invention, there is provided a method of providing a low calorie food composition, said method comprising formulating a food product to contain, at least as a portion of its lipid content, dialkyl ether compounds having the formula



30 wherein R_1 and R_2 are the same or different, saturated or unsaturated, straight or branched, alkyl hydrocarbon chains of from 8 to 30 carbon atoms; and R is either hydrogen or $\begin{array}{c} \text{C} - \text{R}_3 \\ || \\ \text{O} \end{array}$ where R_3 is an acyclic hydrocarbon chain,

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saturated or unsaturated straight or branched, of from 3 to 21 carbon atoms.

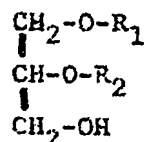
Preferably R_1 and R_2 are alkyl chains of from 12 to 24 carbon atoms and R is hydrogen.

The diethers may be prepared by reacting alkyl alcohols with epihalohydrins in the presence of a Lewis acid. Blends of

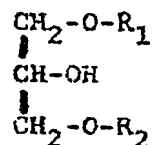
the dialkyl glycerol ethers may be engineered to provide functionality, such as melting characteristics, approximating that of conventional fats.

The crux of the present invention is that the dialkyl glycerol ethers described herein may be selected and/or blended so as to exhibit all the physical and organoleptic properties of conventional fats, i.e., the glycerol triesters. Moreover, unlike conventional fats, the present dialkyl glycerol ethers are not absorbed or digested to any significant degree in the human alimentary tract. Accordingly, the present invention enables the formulation of edible, low-calorie, low-cholesterol food compositions. Such food compositions may be fabricated by combining a protein source material (such as soy protein), a carbohydrate source material (such as starch), and the present dialkyl glycerol ethers. More commonly, however, the present invention will involve providing a food product in which all, or at least a portion, of its conventional fat content has been replaced by dialkyl glycerol ethers. Inasmuch as the diethers are essentially non-absorbable or non-digestible, the resulting food product constitutes a low-caloria, low-cholesterol food composition.

The diether compounds of the present invention preferably have the following formulas:



and



1,2-dialkyl glycerol ether

1,3-dialkyl glycerol ether

wherein R_1 and R_2 may be the same or different, and denote saturated or unsaturated, straight or branched alkyl hydrocarbon chains of from 8 to 30 carbon atoms, and preferably from 12 to 24 carbon atoms. Also, R_1 and R_2 may

be a mixture of different chain lengths, such as C-16 and C-18.

The dialkyl glycerol ethers of the present invention may be made in accordance with various art techniques, and the particular process, or synthesis route utilized, forms no part of the present invention. However, we have found that a synthesis route using monohydric alcohols and epihalohydrins is particularly suitable. The first step in synthesizing the dialkyl glycerol ethers via the epihalohydrin route involves reacting a monohydric alcohol having the formula R_1-OH with an epihalohydrin such as epichlorohydrin or epibromohydrin. The reaction is generally carried out at a temperature of from about 10 to 100°C. at atmospheric pressure, and preferably at from about 50-100°C. Stoichiometric amounts, or molar excesses up to 10:1, of alcohol and epihalohydrin may be utilized. Anhydrous conditions are maintained for best results. The reaction should be carried out in the presence of a catalytic amount of a Lewis acid, preferably stannic chloride. Other Lewis acids that may be utilized include sulfuric acid, hydrofluoric acid, boron trifluoride, ferric chloride, and metal halides.

The alcohol reactant may be any saturated or unsaturated, straight or branched chain, monohydric alkyl alcohol having from 8 to 30 carbon atoms, and preferably from 12 to 24 carbon atoms in the molecule. Preferably, the alcohol reactant is a fatty, unbranched alcohol, either saturated or unsaturated, having from 12 to 24 carbon atoms in the fatty chain. Typical monohydric alcohols that may be utilized in the present reaction include octyl, decyl, dodecyl, tetradecyl, hexadecyl, octadecyl, cis-9-octadecenyl, hydrogenated tallow alcohols, and mixtures thereof. If the particular alcohol reactant is a solid at

room temperature, then the reaction temperature must be sufficient to place the alcohol in a liquid state, or reaction must be carried out in a suitable solvent.

The reaction product obtained by this first synthesis step is a mixture comprising primarily 1-alkoxy-3-chloropropanol-2 (glycerol halohydrin alpha-alkyl ether) along with a minor amount of 2-alkoxy-3-chloropropanol-1 (glycerol halohydrin beta-alkyl ether). Preferably, reaction by-products and unreacted compounds (such as alcohol) present in the reaction mixture are removed at this point by conventional means such as distillation or stripping. However, removal of such by-products and excess reactants is not necessary. The mixture of glycerol halohydrin alkyl ethers is then treated with a concentrated aqueous solution of a strong base, such as sodium hydroxide, resulting in a ring-closing reaction to form the corresponding alkyl glycidyl ethers. The reaction generally proceeds at temperatures of from 0 to 100°C.

Finally, the mixture of 1-alkoxy-2,3-epoxypropane (alkyl glycidyl ethers) is further reacted with a stoichiometric amount or molar excess, preferably from 2:1 to 10:1, of additional monohydric alcohol having the formula R_2-OH , (wherein R_2 is defined in the same manner as R_1), in the presence of a Lewis acid, preferably stannic chloride. The reaction is generally carried out at temperatures of from 10 to 100°C.

The final reaction mixture contains 1, 2-dialkyl glycerol ethers (2,3-dialkoxypropanol-1) and 1,3-dialkyl glycerol ethers (1,3-dialkoxypropanol-2), as well as excess alcohol and various by-products. The glycerol dialkyl ethers generally comprise about 50-55% by weight of the reaction mixture, unreacted alcohol comprising about 5% of

the reaction mixture, and by-products comprising about 40-45% by weight. Preferably, the reaction mixture is treated by conventional means, such as stripping or distillation or treatment with a strong base, to remove excess alcohols and by-products. However, it should be understood that this is not an essential step of the present process. On the contrary, we have discovered that the presence of reaction by-products, which we believe to be polyether materials containing organically bound chlorine, apparently is not deleterious to the low caloric value of the diethers. Moreover, these reaction by-products do not materially detract from the fat-simulating functionality of the diethers.

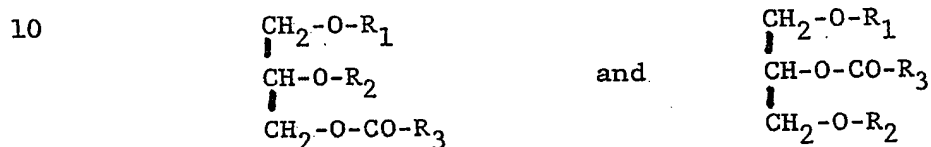
There are several prior art references relating to the preparation of fat substitutes for foods. See U.S. Patents 2,962,419; 3,579,548; and 3,600,186. However, these references are directed to the preparation of various synthetic ester compositions, such as sucrose esters, which are represented to have lower caloric values than conventional fats (glycerol triesters). Although the ester compositions set forth in these references do have lower caloric contents than conventional fats, the caloric content of the food compositions remain substantial, as such esters are apparently capable of absorption within the digestive tract. On the other hand, in the course of rat feeding studies, we have surprisingly observed that the dialkyl glycerol ethers of this invention are absorbed in only very small amounts within the digestive tract. It is believed that this phenomenon of non-absorption is due to the inability of natural enzymes present in the digestive tract to cleave the ether bonds of the dialkyl ether compounds. Moreover, the present diethers, even when combined with natural fats, do not exhibit increased caloric levels.

It is known that glycerol triethers are not readily absorbed in the digestive tract, and that such triethers have structural similarities to natural fats and oils. It has also been suggested that such glycerol triethers could be used to decrease the total caloric value of food products while providing the desired functional characteristics normally contributed by natural fats. See Journal of the American Oil Chemists Society, pages 427-429, October 19, 1975. However, such glycerol triether compounds are extremely
10 difficult and expensive to synthesize, and therefore offer no present commercial possibility as a conventional fat substitute. Of course, glycerol monoethers are commercially available and relatively easy to prepare. However, glycerol monoethers are readily absorbed in the alimentary canal, and therefore offer no substantial reduction in caloric intake.

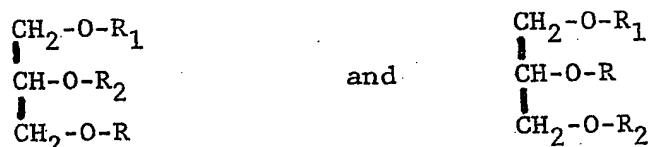
It should be noted that, if desired, the free hydroxyl group of the dialkyl glycerol ethers may be esterified with a fatty acid so as to provide a single ester group in the glycerol diether molecule. It is felt that the single
20 ester group, especially when located in the beta position on the glycerol molecule, is not easily cleaved by natural enzymes in the digestive tract, probably due to steric hindrance provided by the long chain alkyl ether groups. When it is desired to replace the free hydroxyl group in the dialkyl glycerol ether, the diether is merely reacted with a suitable fatty acid under conventional esterification reaction conditions. Suitable fatty acids may be defined by the formula R_3COOH wherein R_3 is an acyclic hydrocarbon chain, saturated or unsaturated, straight or branched, of from
30 about 3 to 21 carbon atoms. Typical fatty acids that may be used include butyric, valeric, caproic, octanoic, decanoic, decenoic, lauric, cis-9-dodecenoic, myristic, cis-

9-tetradecenoic, pentadecenoic, cis-9-pentadecenoic, palmitic, stearic, linoleic, behenic, erucic, eleostearic acids and mixtures thereof. The fatty acids can be derived from naturally occurring or synthetic fatty acids. Also, naturally occurring fats and oils can serve as the fatty acid source, e.g., coconut, palm, palm kernel, babassu, corn, peanut, safflower seed, sesame seed, sunflower seed oils and lard.

The glycerol monoester diether compounds of the present invention have the following formulas:



wherein R_1 , R_2 and R_3 are as defined above. Broadly then, the present invention involves the use of the following compounds as fat substitutes in food products:



wherein R_1 and R_2 are as previously defined, and R is either
 20 a hydrogen atom or -C-R_3 where R_3 is an acyclic hydrocarbon
 O

chain, saturated or unsaturated, straight or branched, of from about 3 to 21 carbon atoms.

It is important to note that although no single dialkyl glycerol ether or glycerol monoester diether prepared in accordance with this invention will likely exhibit all the structural similarities and functionalities of conventional fats, blends of various dialkyl glycerol ethers and/or monoester diethers prepared in accordance with this invention
 30 may be engineered so as to provide a wide range of fat-like characteristics and functionalities. Thus, blends of various dialkyl glycerol ethers may be prepared so as to provide

broad melting curves (no sharp melting point), and may thus be used as shortening compositions or in margarine. On the other hand, blends may be prepared in order to provide a relatively sharp melting curve so as to simulate fats used as frying fats and confectionery fats. The appropriate selection and blending of dialkyl glycerol ethers prepared in accordance with the present invention is well within the skill of those in the fat and oil art, and is therefore not a part of the present invention. Suffice it to say that

10 reference to various analytical tools such as melting points and solid fat index curves will enable those in the art to prepare blends of dialkyl glycerol ethers suitable as substitutes for a wide range of conventional fats and oils.

The following examples are presented to illustrate the invention; it will be understood that these examples are illustrative only, and should not be considered as limiting the invention in any respect.

EXAMPLE I

Preparation of Di-Dodecyl Glycerol Ether

20 Dodecyl alcohol (2144 g, 11.5 mole) was added to a five liter flask equipped with mechanical stirrer, thermometer, dropping funnel, and condenser. The flask was heated by means of an electrical mantle. When the temperature reached 80°C., 8.0 ml anhydrous stannic chloride was added to the alcohol with stirring. The addition of 625 g (6.8 moles) of epichlorohydrin was begun. The temperature was allowed to rise to 90°C. and maintained there throughout the epichlorohydrin addition -three hours. After a further two hours at

30 90°C., a solution of 320 g (8.0 mole) sodium hydroxide in 500 ml water was added. The mixture was then washed with hot water, dried over anhydrous sodium sulfate and passed through a falling-film molecular still to remove volatiles.

The major component of the residue was dodecyl glycidyl ether.

Dodecyl alcohol (930 g, 5 mole) was charged to a five liter reaction flask equipped with mechanical stirrer, thermometer, reflux condenser, and addition funnel. The flask was heated by means of an electrical mantle. When the temperature had reached 85°C., 5 ml anhydrous stannic chloride was added. The addition of 1210 g dodecyl glycidyl ether was begun. The temperature was allowed to increase to 90°C. and maintained there during the dodecyl glycidyl ether addition and for a further three hours. Then 300 ml saturated sodium carbonate solution was added. The mixture was then washed with hot water, and passed through a falling-film molecular still under reduced pressure to remove volatiles. The residue was primarily didodecyl glycerol ether.

EXAMPLE II

Preparation of Cis-9-Octadecenyl Octadecyl Glycerol Ether

Cis-9-Octadecenyl octadecyl glycerol ether was prepared by the procedure of Example I except that 4288 g (16 mole) cis-9-octadecenyl alcohol was reacted with 1250 g (13.5 mole) epichlorohydrin and treated with strong base to form the cis-9-octadecenyl glycidyl ether. Then 1350 g (5 mole) octadecyl alcohol was reacted with 1640 g. of the cis-9-octa decenyl glycidyl ether to form the mixed glycerol diether.

EXAMPLE III

Preparation of Cis-9-Octadecenyl-Tallow Alcohol Glycerol Ether

Hydrogenated tallow alcohols (1350 g, 5 mole) were reacted with 1640 g cis-9-octadecenyl glycidyl ether (prepared as in Example II) using the procedures outlined in Example

I. The product was the mixed glycerol diethers.

E X A M P L E IV

Preparation of Dioctadecyl Glycerol Ether

Dioctadecyl glycerol ether was prepared by the procedure of Example I except that 4320 g (16 mole) octadecyl alcohol was reacted with 1472 g (16 mole) epichlorohydrin in the formation of the octadecyl glycidyl ether. Then 1350 g (5 mole) octadecyl alcohol was reacted with 1495 g octadecyl glycidyl ether to form the dioctadecyl glycerol ether.

10

E X A M P L E V

A blend was formulated from the reaction products prepared in accordance with Examples I-IV, which blend had the following composition:

<u>COMPONENT</u>	<u>WEIGHT %</u>
Reaction Product of EX. I (primarily didodecyl glycerol ether)	20
Reaction Product of EX. II (primarily cis-9-Octadecenyl octadecyl glycerol ether)	35
Reaction Product of EX. III (primarily cis-9-octadecenyl-tallow alcohol glycerol ether)	35
Reaction Product of EX. IV (primarily dioctadecyl glycerol ether)	10

20

This blend was then bleached with an acid-activated bleaching earth, and subsequently steam deodorized. The blend had solid fat index values of 53.6 at 50°F., 23.5 at 70°F., 19.9 at 80°F., 13.8 at 92°F., and 8.8 at 100°F. The blend had nuclear magnetic resonance (NMR) values of 58.8 at 10°C., 30.1 at 20°C., 16.0 at 30°C., and 6.2 at 40°C. Also, the blend exhibited a Mettler dropping point of 43.8°C.

E X A M P L E VI

30

Butter cookies were prepared by combining 113 grams sugar, 5.5 grams salt, 75 grams whole egg, 227 grams cake flour, 3 drops flavor, and either 184 grams pure all

purpose shortening or 184 grams of the blend of Example 5. The sugar, shortening or diether substitute, and salt were blended in a mixer for five minutes at medium speed. Whole egg was added with mixing continued for another 3 minutes. Flour was added and the mixture was blended an additional 30 seconds. The dough was then extruded into cookies and baked for 9 minutes at 375°F. Light, buttery flavor cookies resulted when either the all purpose shortening or the blend of dialkyl glycerol ethers was used.

10

E X A M P L E VII

Butter cream icing was prepared by blending 227 grams sugar, 70.8 grams all purpose shortening or the blend of Example V, 28.4 grams water, 14 grams non-fat dry milk, 1.4 grams emulsifier (used with di-alkyl glycerol ether blend only), 1 gram salt and 1 gram vanilla. All of the ingredients were creamed in a mixer at medium speed for 12 minutes. A white creamy icing resulted when either regular all purpose shortening or the dialkyl glycerol ether blend was used.

20

E X A M P L E VIII

Pie crust was prepared by mixing the following for 40 seconds: 75 grams of flour and 112.5 grams of either pure all purpose shortening or the blend of Example V. Thereafter, 75 grams additional flour were added and the mixture was blended for another 15 seconds. 45 grams ice water, 3 grams corn sugar and 5.3 grams salt were then added with mixing for 15 seconds. The dough was rolled and pie shells were prepared. They were baked for 10 minutes. The crusts were light brown and flaky. The crust obtained using the dialkyl glycerol ether blend of Example V was indistinguishable from that made from the pure all purpose shortening.

30

E X A M P L E IX

Margarine was prepared by combining 272 grams of the blend of Example V with 63 grams water, 8.6 grams salt, 0.86 grams lecithin, 0.05 grams coloring and 0.05 grams flavor.. All of the ingredients were heated in a jacketed blender to about 60°C. The mixture was then cooled as quickly as possible with continuous mixing. When the mixture was in a semi-solid state, it was poured into a container and allowed to harden at about 4°C. A hard margarine resulted.

10

E X A M P L E X

Using food energy values found in the USDA Composition of Foods, Agriculture Handbook No. 8, the following caloric values and percentage reductions can be estimated:

<u>Food Product</u>	<u>Calories per 100 Grams of All Purpose Shortening - Control</u>	<u>Calories per 100 Grams of Dialkyl Glycerol Ether Blend</u>	<u>% Calories of Control</u>
Butter cookie	499	229	46%
Butter cream icing	451	269	60% Pie
crust	573	206	36%
20 Margarine	697	0	0

E X A M P L E XI

13 grams of frozen French fried potatoes (ripple cut) were fried in 250 grams of all purpose shortening. Also, 13 grams of frozen French fried potatoes were fried in 250 grams of the blend of Example V. Frying times for both tests were 3 minutes at 375°F. Typical tasting and looking French fried potatoes resulted from both tests. The dialkyl glycerol ether blend produced essentially identical results as the all purpose shortening.

30

E X A M P L E XII

Preparation of Dihexadecyl Glycerol Ether

Hexadecyl alcohol, 1225 g (5.05 moles), was placed

in a five liter reaction flask equipped with dropping funnel, mechanical stirrer, reflux condenser, and thermometer. The reflux condenser was fitted with a drying tube for restricting entrance of moisture. An electrical mantle was used for heating. Heat was applied to the flask and, when the temperature of the alcohol had reached 80°C., the stirrer was started and 5.0 ml (11 g, .043 moles) stannic chloride was added. After a few moments the addition of 462.5 (5.00 moles) of epichlorohydrin was begun. The epichlorohydrin addition required 3-1/2 hours. The reaction temperature was maintained at 85-90°C. during the epichlorohydrin addition and for a further 1-1/2 hours. The resultant hexadecyl glycerol halohydrin ether was not isolated. To the reaction mixture was added a solution of 202 g (5.05 mole) sodium hydroxide in one liter water. The mixture was stirred under reflux for 8 hours. The reaction mixture was washed with hot water, and then the residual moisture and volatiles were removed by passing the product through a wiped-film molecular still under reduced pressure. The hexadecyl glycidyl ether product weighed 1063 g and analyzed for 4.55% oxirane oxygen (theoretical: 5.36%).

Dihexadecyl glycerol ether was prepared as follows:
Hexadecyl alcohol, 243 g (1 mole), was added to a one liter reaction flask equipped with mechanical stirrer, dropping funnel, reflux condenser, and thermometer. The condenser was fitted with a drying tube. The flask was heated with an electrical mantle. When the alcohol temperature had reached about 80°C., 1 ml (2.2 g, .0085 moles) stannic chloride was added. Then the addition of 352 g hexadecyl glycidyl ether (weight equivalent to one mole) was begun. The addition required about 4 hours. The temperature was allowed to rise to about 90°C. and maintained

there during the hexadecyl glycidyl ether addition and for a further 1-1/2 hours. A solution of 10 g sodium carbonate in 40 ml water was added and then the reaction mixture was washed with hot water. The oily product was filtered and then volatiles were removed by passage through a wiped-film molecular still under reduced pressure. The dihexadecyl glycerol ether product weighed 502 g.

E X A M P L E XIII

Preparation of Dioleoyl Glycerol Ether

10 Oleyl glycidyl ether (cis-9-octadecenyl-2,3-epoxypropane) was prepared by the procedure of Example XII except that 537 g (2 mole) oleyl alcohol was reacted with 185 g (2 mole) epichlorohydrin using 2.0 ml anhydrous stannic chloride. The product weighed 386 g and had an oxirane oxygen content of 4.48% (theoretical 4.93%). Dioleoyl glycerol ether was then prepared from 242 g (0.9 mole) oleyl alcohol and 321 g of the above oleyl glycidyl ether. The product weighed 411 g and consisted mainly of dioleoyl glycerol ether.

20 The above experiments indicate that a dialkyl glycerol ether blend can provide fat-like functionality in many different food applications. This can be done without introducing unpalatable flavors or textures. Thus, the present dialkyl glycerol ethers may be incorporated in either formulated food products or in food products in which a partial amount of the natural fat content has been removed.

 Obviously, any one blend is not the best for each and every application. Best results can be obtained using blends of dialkyl glycerol ethers customized to the product.

30 Therefore, it should be clear that the above experiments are not intended to exhaust the possibilities for substituting dialkyl glycerol ether blends in lieu of fats and oils in

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all known food applications. Those skilled in the art of fat and food technology will envision other uses not described here.

10 Rat feeding studies were conducted using male Holtzman white albino rats. A basal diet was established comprising commercial rat food at a level of 0.12 grams per gram live weight on a daily basis, adjusted three times weekly for changes in body weight. For some rats in the study, the basal diet was modified by substituting 10% by weight of the commercial feed with blends of dialkyl glycerol ethers. Test results showed that the dialkyl glycerol ethers were generally non-digestible in the alimentary tract, indicating extremely low caloric values.

Obviously many modifications and variations of the invention as hereinbefore set forth may be made without departing from the spirit and scope thereof, and, therefore, only such limitations should be imposed as are indicated in the appended claims.

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